

Normalization references for Europe and North America for application with USEtox™ characterization factors

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Abstract

Purpose In life cycle impact assessment, normalization can be a very effective tool for the life cycle assessment practitioner to interpret results and put them into perspective. The paper presents normalization references for the recently developed USEtox™ model, which aims at calculating globally applicable characterization factors. Normalization references for Europe and North America are determined, and guidance for expansions to other geographical regions is provided.

Materials and methods The base years of the European and North American inventories are 2004 and 2002/2008, respectively. Emission data were extracted from two literature sources referring to each of the considered regions. The inventory for North America was adapted to avoid extrapolation of data from other regions and thus bring consistency with the emission inventory for Europe. In spite of different inventory assumptions, a similar coverage of substances was obtained for both regions with relatively high representation of metals and a number of organic compounds, mainly consisting of non-methane

volatile organic compounds and pesticides. The two inventory sets were eventually characterized with the characterization factors (CFs) calculated with the version 1.0 of the USEtox™ model and substance database; both interim and recommended CFs were used.

Results and discussion Normalization references are provided for Europe and North America for the three USEtox™ toxic impact categories; ratios between the normalization references for the two regions in all cases lie below a factor of 3. Causes for the observed discrepancies are found to be different inventory assumptions as well as variations in the type and intensity of actual emissions between the two regions. Additional causes are inventories that only cover a limited number of substances, and the characterization model, which can only provide interim factors for certain substances like metal compounds. Based on these causes and on a review of recent studies on normalization references, a list of substances to be prioritized when collecting emission data was built, demonstrating the importance of metals.

Conclusions In the perspective of further refining the presented normalization references and of calculating new references for other regions, guidance is provided including a list of priority substances that should be considered when building emission inventories for normalization references.

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1 Introduction

Normalization is a step in the interpretation of results of a life cycle assessment (LCA) study. Defined as optional in

the ISO 14044:2006 (ISO 2006), normalization can be useful for the LCA practitioner when providing recommendations to decision stakeholders with regard to environmental sustainability. The characterized results from a given LCA are scaled against a reference—the normalization reference—allowing quantification of the magnitude of each impact relative to the reference as well as enabling comparison across impact categories, which inherently implies weighting (either with equal weights if no weighting factors are applied, or using specific weighting factors per impact category). The normalization references can be considered as the characterized results of a “large scale” system LCA, in which the inventory analysis covers the background load of the society’s activities within a certain region and in a certain reference year. Two different approaches exist to define the normalization references, either as aggregates (e.g., CML; Guinée et al. 2002) or as per-capita impacts (e.g., EDIP; Wenzel et al. 1997). While the former leads to normalized impact scores describing the share of impacts from the assessed product system in the total environmental impacts within the reference region, the latter may offer the advantage to be more communicative due to its focus on the impact from an individual, all normalized impact scores being expressed as person equivalents, i.e. the annual contribution of an average person in the defined area (Wenzel et al. 1997; Guinée et al. 2002).

A need for consensus regarding toxicity-related impact modeling, which still suffers from large uncertainties, has led to the recent launch of the USEtox™ model, which was developed as part of the Life Cycle Initiative by the United Nations Environment Programme and the Society of Environmental Toxicology and Chemistry. USEtox™ is a consensus model, which resulted from the review of the most relevant toxic impact assessment models commonly used in LCA methodologies (Rosenbaum et al. 2008). It currently enables the calculation of globally applicable characterization factors for assessing three impact categories at midpoint, namely freshwater ecotoxicity and human toxicity, differentiated into cancer effects and non-cancer effects. Although it is not intended to supersede the other toxicity-related life cycle impact assessment (LCIA) methods, the USEtox™ model is aimed to be a globally applicable model. USEtox™ is considered by the European Commission as a possible recommended midpoint characterization model for freshwater ecotoxicity and human toxicity impact categories, and it is expected to be incorporated as the characterization model for human and ecotoxic impacts in the North American LCIA methodology TRACI. It is furthermore under consideration for adoption by several national and international governmental organizations and bodies and thus likely to become a reference toxic impact assessment

model into which further developments are to be integrated, e.g., inclusion of an indoor exposure module (Hellweg et al. 2009). In this context, there is a need for normalization references for the USEtox™-based impact categories in order to interpret LCA results in a comprehensive manner.

The paper provides sets of per-capita normalization references for the three USEtox™ impact categories for both Europe and North America, two regions for which inventory data are sufficiently available. Because of the global dimension associated with the model and because the geographical scope of product LCA is often stretched over the world, we also address the need to expand the calculation of normalization references to other economic systems, e.g., other continents or the world as a whole. Guidance on how to conduct such extensions in connection to the inventory analysis is also provided.

2 Materials and methods

2.1 Inventory for Europe (EU)

The inventory for Europe was extracted from Laurent et al. (2011); it covers 38 European countries. Laurent et al. (2011) collected emission data for the reference year 2004 from different databases and monitoring bodies, complemented by extrapolations where necessary. Estimates based on extrapolations of emission data from regions outside Europe were avoided because of potential differences in the society’s activities between regions (see Section 4.2). In the inventory settings, emphasis was put on the differentiation of non-methane volatile organic compounds (NMVOC) into individual compounds and on an appropriate modeling of pesticide emissions as well as on reporting heavy metals in the most comprehensive way. Although briefly recalled below, information on those activities are further detailed in Laurent et al. (2011).

NMVOC emissions were differentiated into single compound emissions (specification for 318 different chemicals) according to emission sources, each source type being assigned a typical chemical composition. The combination of the obtained matrix with the corresponding USEtox™ characterization factors (CFs), when available, enabled the calculation of emission-weighted generic factors for characterizing NMVOC for 80 emission sources (see [Electronic Supplementary Material](#)). This can provide additional support to the LCA practitioner when available data are not sufficient to allow differentiation of NMVOC, and it can serve to identify additional substances to be characterized by the USEtox™ model in the future (those suspected to be toxic and/or having a large share in the distribution profiles; not shown here).

Similarly to NMVOC, pesticides were differentiated into 482 active ingredients, and their releases were estimated based on consumption data. In the present study, emissions of pesticides were modeled following two different approaches, thus leading to two separate sets of normalization references for Europe—see Section 3. The first approach follows guidance from Laurent et al. (2011) and implies the building of the inventory using the PestLCI model (Birkved and Hauschild 2006). Considering that pesticides are intentionally applied on the agricultural field, which should be considered part of the technosphere, only the fraction reaching the ecosphere is included in the inventory with this approach. Averaging results from PestLCI, this fraction is estimated to consist of 5% of applied pesticides as direct emissions to air (e.g., from wind drift) and 0.1% of applied pesticides as direct emissions to water (e.g., through run-off). Because the PestLCI model has not been widely applied until now, the traditional approach, consisting of inventorying 100% of the consumed pesticides as direct emissions to agricultural soil, was additionally included in the present study; this second approach is referred to as “Pest-to-soil” in the paper. The question of which approach to consider in practice falls outside the scope of this study and is, therefore, not addressed here; for such discussion, the reader is referred to Laurent et al. (2011).

With respect to heavy metals, the availability of the emission data is strongly dependent on the emission compartment. Air-borne emissions are relatively well covered for a number of metals by the EMEP Centre (<http://www.ceip.at/>). Water-borne releases are reported in a less comprehensive manner: typically, monitoring bodies (e.g., OSPAR Commission 2006) report inputs of most common metals to certain seas (copper, zinc, mercury, cadmium, lead), from which emissions to freshwater can be derived (see Laurent et al. 2011). A number of other relevant heavy metal releases that are missing in these reports (e.g., iron or chromium) were added, based on emission factors from energy production (Ecoinvent Centre 2007). Heavy metals emitted to soil could only be estimated via their contents in sewage sludge applied to agricultural fields (emissions to industrial soil not considered).

2.2 Inventory for North America

To quantify emissions for North America, the life cycle inventory by Lautier et al. (2010) was used and slightly adapted to bring consistency with the EU inventory. Their inventory covers the USA and Canada, thereby excluding Mexico. The following paragraphs provide an overview of the main points describing the NA inventory, while for a detailed description, the reader is referred to Lautier et al. (2010).

The US inventory consists of updated data from Bare et al. (2006) covering industrial, commercial, residential, and agricultural activities, as well as energy production and transport for the reference year 2008, except for pesticides where it is 2002. The main data sources are the National Emission Inventory (NEI; emissions into the air of 353 Hazardous Air Pollutants), and the Toxic Release Inventory (for emissions into air, water, and soil of hazardous substances not covered by the NEI). The pesticide emissions are taken from USEPA (Kiely et al. 2004). Similarly to the inventory for Europe, two approaches were considered for modeling pesticide releases, i.e., the “Pest-to-soil” approach, used in Lautier et al. (2010), and the PestLCI approach—see Section 2.1. This also implies two separate sets of normalization factors for North America—see Section 3. For emissions of metals into soil, the emissions of metals and metal compounds are typically reported separately. Since the characterization factors for metals would overestimate the impact of metal compounds, the emissions of metal compounds into soil were excluded in the present NA inventory.

The Canadian emission inventory contains data for 299 substances covering industrial, commercial, residential, and agricultural activities, as well as energy production and transport for the reference year 2005. The main data sources are the inventories compiled by Environment Canada, which publishes the National Pollutant Release Inventory based on the declarations of facilities that exceed a certain criteria. For criteria air contaminants, the inventory is exhaustive in terms of emission sources. As no data are available for pesticide emissions in Canada, data gaps were filled with extrapolated data from the US inventory from USEPA (Kiely et al. 2004). The extrapolations are based on crop-production-area for insecticides and on gross domestic products (GDP) for metal emissions (reported as “metals and compounds” and thus impossible to be differentiated into single metals), herbicides, fungicides, and other pesticides as recommended by Sleeswijk et al. (2008). Lautier et al. (2010) filled data gaps related to Canadian emissions of metals into agricultural soil (metal content in pesticides and fertilizers) with extrapolated values from the European inventory of Sleeswijk et al. (2008). However, in the present study, those were removed because the used extrapolation approach is believed to lead to overestimations for North America as well as it would add inconsistencies with the EU inventory. Therefore, Canadian metal emissions into soil only cover industrial sources.

2.3 USEtox™-based characterization factors and impact categories

The USEtox™-based characterization factors used to calculate the normalization references for the three toxic

impact categories were taken from the version 1.0 of the model (dating February 2010). Three emission compartments are modeled—air, water, soil—further divided in six sub-compartments, viz. continental air, urban air, continental freshwater, sea water, agricultural soil, and natural soil. Because air-borne emissions from the inventories of both Europe and North America are not further specified, they were coupled with averaged characterization factors for releases to continental air and urban air. Although the impact of this assumption has not been investigated in the present study, future updates should aim to reduce the introduced bias by consistently allocating air-borne releases to either continental or urban air emissions..

Some of the USEtox™ characterization factors are classified as interim due to the quality and appropriateness of the model for this type of chemicals (e.g., the metal compounds and dissociating organic chemicals) or of the substance data used for calculating the characterization factors (see Rosenbaum et al. 2008). Both interim and recommended characterization factors were used in the calculations of the normalization references. The main purpose was to cover as many inventoried substances as possible even though uncertainties increase—see Section 6.1.

3 Results

The per-capita normalization references for the three toxic impact categories are provided for North America and Europe in Table 1. Where relevant, results are differentiated according to the type of pesticide modeling, either the PestLCI model or the “Pest-to-soil” approach (see Section 2.1). Ratios between normalization references in both regions are also presented.

The normalization references provided in Table 1 show the calculated per-capita impacts for Europe and North America within the reference years. The normalization references for human toxicity (cancer effects) represent the number of cancer cases caused by the emissions assigned to an average person in the reference year. The results in Table 1 mean that the emissions associated with the activities of app. 10000 (North America) or 31000 (Europe) persons in the reference year will lead to one incident of cancer. For non-cancer effects, the figures are higher: one incident resulted from the annual emissions associated with app. 1230 (Europe) or 2200 (North America) persons in the reference year. For freshwater ecotoxicity the figures represent the product of the fraction of species affected above their EC50 value (the concentration affecting 50% of the individuals in the test), the volume within which this exposure took place and the duration. The values in Table 1 mean that the chemical emissions from an average person could lead to exposure of all species in the ecosystem above

their EC50 value for the duration of a year for, respectively, app. 5,000–8,800 m³ freshwater (Europe) or 5,100–14,700 m³ freshwater (North America).

These figures can be put in perspective with regional statistics for Europe and North America. The volume of water in river channels, freshwater lakes, and inland salt lakes are estimated to be app. 80,100 and 25,892 km³ in Europe and North America, respectively (Shiklomanov and Rodda 2003).¹ Normalized by the population in the two regions, the above figures would thus correspond to $1.1 \cdot 10^5$ m³ (Europe) and $7.7 \cdot 10^4$ m³ (North America) per person. According to the figures in Table 1, chemical emissions for an average person could thus lead to exposure of all species in the ecosystem above their EC50 value for the duration of a year in ca. 5–8% and 7–19% of the total volume of inland water flowing in Europe and North America.

With respect to human toxicity, numbers of cases for non-cancer effects are typically hard to obtain due to the very broad scope of the effects and the difficulty in monitoring them in a comprehensive manner. However, cancer cases are better reported. Ferlay et al. (2010) estimated that 1,604,000 and 3,213,000 new cancer cases were expected to be diagnosed in 2008 in North America and Europe, respectively. Those numbers correspond to one case out of 208 and 224 for North America and Europe, respectively. It thus indicates that ca. 47 times more cancer cases than what is reflected by the normalization reference were expected to be diagnosed in North America; this figure goes up to ca. 138 when comparing situations in Europe. This suggests that the fraction of all cancer incidents caused by chemical exposure is limited. This finding is in agreement with the etiology of cancer incidence conducted by the WHO, which report environmental pollution and occupational exposures to chemicals as causes much less contributing to the cancer's burden than other sources like tobacco or radiations (IARC 2008).

4 Discussion

4.1 Comparisons of the two regions

Table 1 shows that the per-capita normalization references for Europe and North America fall within the same order of magnitude, with ratios of ca. 1–3. The distribution of the contributing chemicals to the impact potentials can however differ between the two sets of results, which may render

¹ Although neglected here, certain salt lakes are endorheic basins with no outflow of water except evaporation, hence incompatible with the inventory analysis behind the calculated normalization references; because of limited data availability on the proportions of such cases in the total lake basins, this incompatibility was, however, disregarded.

Table 1 Normalization references for USEtox™ for Europe and North America

USEtox™-based impact categories	Pesticide modeling ^a	Europe	North America	Ratio ^b
Freshwater ecotoxicity (CTU _e /person/year)	Pest-to-soil	$8.72 \cdot 10^3$	$1.47 \cdot 10^4$	1.7
Freshwater ecotoxicity (CTU _e /person/year)	PestLCI	$5.06 \cdot 10^3$	$5.12 \cdot 10^3$	1.0
Human toxicity, cancer effects (CTU _h /person/year)	PestLCI/Pest-to-soil ^c	$3.25 \cdot 10^{-5}$	$1.02 \cdot 10^{-4}$	3.1
Human toxicity, non-cancer effects (CTU _h /person/year)	PestLCI/Pest-to-soil ^c	$8.14 \cdot 10^{-4}$	$4.54 \cdot 10^{-4}$	0.6

CTU_e comparative toxic unit for ecosystems, expressing an estimate of the potentially affected fraction of species integrated over time and volume per unit mass of a chemical emitted (PAF m³ year), CTU_h comparative toxic unit for humans, expressing the estimated increase in morbidity in the total human population per unit mass of a chemical emitted (cases)

^a Normalization references provided for the two different approaches for inventorying pesticides: “Pest-to-soil” referring to 100% of applied pesticides directly emitted to agricultural soil, and “PestLCI” assuming 5% and 0.1% of applied pesticides directly emitted to air and water, respectively (no emissions to soil)—see Section 2 for further details

^b Ratio between normalization references {North America}/{Europe}

^c Influence of pesticide inventory modeling is negligible on normalization references for human toxicity (maximum difference <1%)

this apparent agreement coincidental. A contribution analysis was thus conducted in order to identify the substances, which contribute the most to the different impact categories (Table 2).

As visible from the contribution analysis, a limited number of chemicals drive the normalization references, regardless of the considered reference region (the gray cells in Table 2). Excluding freshwater ecotoxicity for North America, for which the chemical contributions are more broadly distributed, it is indeed noteworthy that less than five substances govern more than 70% of the impact potentials for both regions. This is likely to result from incompleteness in the inventory, as several thousands of potentially harmful chemicals are not covered, as well as from the characterization model, which tends to overestimate the CFs of certain chemicals (e.g., metals)—see Section 6.1.

Overall, a relatively good correlation in the distributions is observed between European and North American results. Although differences exist between the two regions with regard to the magnitude of each substance contribution, most influential substances tend to match. With respect to freshwater ecotoxicity, Table 2 shows that metals and pesticides dominate the normalization references although their respective shares of the impact potentials are strongly dependent on the type of pesticide modeling adopted in the inventory analysis (see Section 2). The high importance of metals in the results suggests that the inventory of their releases, particularly copper and zinc emitted to water (accounting altogether for 44–77% and 13–40% in European and North American normalization references, respectively) are likely to be the major source of the observed discrepancies between the two regions in the contribution analysis. Emission data of copper and zinc in Europe originate from environmental measurements carried out by monitoring bodies whereas the industry data from North America may underestimate the water-borne emissions of

metals due to an incomplete coverage of industrial activities. The strong dependence of the normalization references on metal emissions is also reflected in the contribution analyses for human toxic impacts, all of them showing metals accounting for more than 55% of the impact potentials.

4.2 Causes of variations between the two regions

Difficulties arise when trying to distinguish the causes of variations between the normalization references for Europe and North America, i.e. whether the observed discrepancies are related to different inventory assumptions and data availability or to differences in the actual emission intensities. Laurent et al. (2011) argue that normalization references for toxicity-related impacts are more sensitive to the inventory settings (e.g. coverage of substances) than to actual changes in emissions, but this statement refers to changes over time within the same economic region. Lautier et al. (2010) however compared normalization references between North America and European. Data gaps and inventory assumptions, especially with regard to metal emissions, are identified as significant sources of uncertainties, which may threaten the entire reliability of such comparisons.

Nevertheless, Lautier et al. (2010) also show that differences in the type of industrial activities between regions can substantially influence the normalization references. In the present study, the prominence of arsenic emissions to soil in the normalization references for human toxicity for North America (see Table 2) illustrates this point as emissions of arsenic primarily stem from the US metal mining sector. Because metal mining in Europe is not as intensive as in North America, lower emissions of heavy metals to soil, and probably to other compartments (e.g., freshwater environments) can be expected. Considering the toxicity of many metals, this tends to result in normaliza-

Table 2 Contribution analyses of the USEtox™ normalization references for Europe and North America^a

Substance	Emission compartment	Freshwater ecotoxicity		Freshwater ecotoxicity		Human toxicity (cancer)		Human toxicity (non-cancer)	
		<i>PestLCI</i>		<i>Pest-to-soil</i>		<i>PestLCI/Pest-to-soil</i> ^b		<i>PestLCI/Pest-to-soil</i> ^b	
		Europe	North America	Europe	North America	Europe	North America	Europe	North America
Acrolein	Air	-	-	-	-	-	-	2%	13%
Antimony	Fresh water	1%	-	1%	-	-	-	-	-
Antimony	Soil	-	4%	-	2%	-	-	-	-
Arsenic	Air	-	-	-	-	-	-	-	3%
Arsenic	Fresh water	2%	-	1%	-	3%	-	8%	-
Arsenic	Soil	-	3%	-	1%	-	1%	-	21%
Atrazine	Agricultural soil	-	-	-	9%	-	-	-	-
Barium	Fresh water	-	3%	-	1%	-	-	-	2%
Cadmium	Air	-	-	-	-	-	-	2%	3%
Chlorothalonil	Agricultural soil	-	1%	1%	7%	-	-	-	-
Chlorpyrifos	Fresh water	1%	2%	-	-	-	-	-	-
Chlorpyrifos	Agricultural soil	-	-	11%	16%	-	-	-	-
Chromium	Air	-	-	-	-	6%	3%	-	-
Chromium	Agricultural soil	-	10% ^c	-	3% ^c	2%	50% ^c	-	-
Chromium	Water	-	-	-	-	-	1%	-	-
Chromium VI	Fresh water	3%	6%	2%	2%	42%	32%	-	-
Copper	Fresh water	20%	3%	11%	1%	-	-	-	-
Copper	Air	2%	1%	1%	-	-	-	-	-
Copper	Agricultural soil	1%	19%	-	7% ^c	-	-	-	-
Cyanazine	Agricultural soil	-	-	-	4%	-	-	-	-
Cyfluthrin	Air	4%	3%	-	-	-	-	-	-
Cyfluthrin	Fresh water	3%	2%	-	-	-	-	-	-
Cyfluthrin	Agricultural soil	-	-	3%	1%	-	-	-	-
Cypermethrin	Fresh water	2%	-	-	-	-	-	-	-
Cypermethrin	Air	1%	-	-	-	-	-	-	-
Cypermethrin	Agricultural soil	-	-	1%	-	-	-	-	-
Folpet	Agricultural soil	-	-	21%	-	-	-	-	-
Formaldehyde	Air	-	-	-	-	37%	9%	-	-
Furane	Air	-	-	-	-	2%	-	-	-
Isoproturon	Agricultural soil	-	-	3%	-	-	-	-	-
λ-cyhalothrin	Agricultural soil	-	1%	-	-	-	-	-	-
Lead	Air	-	-	-	-	-	-	6%	9%
Lead	Agricultural soil	-	-	-	-	-	-	1%	-
Mercury	Agricultural soil	-	-	-	-	-	-	4%	-
Mercury	Air	-	-	-	-	4%	1%	19%	25%
Nickel	Soil	-	1%	-	-	-	-	-	-
Phosmet	Agricultural soil	-	-	-	8%	-	-	-	-
Profenofos	Agricultural soil	-	-	-	5%	-	-	-	-
Propanil	Agricultural soil	-	-	-	4%	-	-	-	-
Terbufos	Agricultural soil	-	-	-	4%	-	-	-	-
Terbuthylazine	Agricultural soil	-	-	1%	-	-	-	-	-
Vanadium	Air	-	1%	-	-	-	-	-	-
Vanadium	Soil	-	7%	-	2%	-	-	-	-
Zinc	Fresh water	49%	-	28%	-	-	-	10%	-
Zinc	Air	3%	1%	2%	-	-	-	19%	12%
Zinc	Agricultural soil	2%	16% ^c	1%	6% ^c	-	-	27%	6% ^c
Pesticides	Aggregate	11%	10%	41%	57%	0%	0%	0%	0%
Metals	Aggregate	83%	76%	48%	25%	56%	88%	96%	81%
NMVOC	Air	0%	0%	0%	0%	39%	9%	2%	13%
TOTAL		94%	85%	89%	82%	95%	97%	98%	93%

^a Cut-off of the contributions at 1%; contributions of 10–30% in light gray; contributions above 30% in dark gray^b For human toxicity related impacts, the two pesticide emission modeling approaches (PestLCI or “Pest-to-soil”) lead to the same distribution^c Emissions of chromium, copper, and zinc include releases to both industrial and agricultural soils for North America

tion references for Europe that are lower than those for North America.

Another source of discrepancies between both regions arises from regulations, which may lead to the use and potential release of certain chemicals in one region while they are banned in the other. Uses of specific active ingredients in agriculture are particularly impacted by such differences. Atrazine for example is shown in Table 2 to contribute by 9% to the freshwater ecotoxicity potential in North America whereas it has been banned in the European Union since 2004 and, hence, does not appear in the European inventory.

5 Towards normalization references for other economic systems

The USEtox™ model is aimed to be a globally applicable model. In the absence of information for other regions than Europe and North America, it is advocated to use as a default the set of European normalization references in order to assess product life cycles expanding over several regions; lower variability ranges with respect to inventory modeling were found to be associated with European references—see Section 6.2. However, this temporary alternative should not exclude the need to calculate normalization references for other regions than Europe and North America (other continents, world). Guidance for such purposes is provided below.

Based on the experiences for determining normalization references for Europe and North America, a list of prioritized steps was developed in order to minimize the uncertainties and inconsistencies in the building of the emission inventory. It is recommended (1) to maximize the use of reported emission data, along with proper quality assurance; (2) to prefer extrapolations within the considered region to guarantee similar applied regulations and types of industrial activities (see Section 4.2); (3) to only extrapolate from other economic systems as a last resort, using appropriate parameters (e.g., GDP).

In all three steps, emphasis must be placed on substances, which are likely to influence the magnitude of the normalization references. The contribution analyses provided in Table 2 provide a first indication of chemicals to focus on. However, to gain comprehensiveness, it was decided to broaden the scope and analyze all relevant previous studies, which addressed the determination of normalization references for toxicity-related impacts.

5.1 Top contributing substances in previous studies

Existing contribution analyses for the calculation of normalization references were reviewed, considering studies based

on emission years after 2000. This led to the inclusion of two inventories for Europe: the one used in the present study (Laurent et al. 2011) and the one from Sleeswijk et al. (2008); emission year is 2004 for the former and 2000 for the latter. For North America only the inventory used in the present study was considered (Lautier et al. 2010), referring to emission years 2002/2008. Finally, an inventory for Australia, with emission years 2002/2003, was also included (Lundie et al. 2007). Different LCIA methods were applied in the respective studies from where the contribution analyses were extracted. They comprise USEtox™ (present study), EDIP, ReCiPe, IMPACT2002+, TRACI, and USES-LCA 2.0.

From these studies, the strongest contributing substances across both ecotoxicity-related and human toxicity impact categories were identified for Europe (Table 3) as well as for North America and Australia (Table 4).

5.2 Identification of high priority substances

Although outside the scope of this study, Tables 3 and 4 can serve as a basis for refining the inventories in the regions, which they represent (e.g., by investigating discrepancies within the same region). Here, the eight sets of combined inventories/LCIA methods are analyzed aiming to establish a list of high-priority substances for the calculation of normalization references, regardless of the LCIA method or the region considered. Occurrences of each substance were thus counted in the eight lists detailed in Tables 3 and 4, and all substances present more than twice were deemed of high priority. The resulting list of high-priority substances is provided in Table 5.

As seen in Table 5, metals are dominating among the most recurring chemicals in the contribution analyses (e.g., copper and lead being always reported) and thus constitute the central data on which efforts should be concentrated to collect reliable emission quantities. Although variations exist in the emission intensities, metal releases to the environment, either to air, soil, or water, are likely to occur in any region considered. With respect to the uncertainties related to metal emissions (see Section 2), the provided list can thus serve as a basis to identify which specific metals required to be better monitored and inventoried in the region where the normalization references are intended to be calculated. Along with metals, a number of organics are also to be considered. These chemicals can be classified in three categories, i.e. pesticides, NMVOC, and other organics (e.g. PAH, dioxins). While the last two would always be present, pesticides are more subject to regional regulations (see Section 4.2). The active ingredients provided in Table 5 are, therefore, recommended to be checked and adapted to regional conditions.

Table 3 Top contributing substances in the calculation of normalization references for Europe

USEtox™ (EU-1) The present study	EDIP (EU-1) Laurent et al. (2011)	IMPACT2002+ (EU-2a) Lautier et al. (2010)	ReCiPe (EU-2b) Sleeswijk et al. (2008)
Acrolein	1,3-butadiene	Arsenic	Aldicarb
Antimony	Acetone	Atrazine	Arsenic
Arsenic	Acrolein	Benzene	Atrazine
Cadmium	Benzo(a)pyrene	Benzo(a)pyrene	Barium
Chlorothalonil	Benzaldehyde	Cadmium	Cadmium
Chlorpyrifos	Benzene	Chromium	Chlorine
Chromium	Butanol	Copper	Chloropicrin
Chromium VI	Chlorpyrifos	Decabromophenyl ether	Chlorpyrifos
Copper	Copper	Dioxins	Copper
Cyfluthrin	Cypermethrin	Lead	Cypermethrin
Cypermethrin	Dioxins	Nickel	Lead
Folpet	Ethene	Phenol, pentachloro-	Manganese
Formaldehyde	Formaldehyde	Zinc	Mercury
Furane	Hexanes		Lauryldimethylamine- <i>N</i> -oxide
Isoproturon	Iron		Nickel
Lead	Lead		Selenium
Mercury	Mercury		Terbufos
Terbuthylazine	Parathion-methyl		Vanadium
Zinc	PAH (as B(a)P eq) ^a		Zinc
	Strontium		
	Thallium		
	Vinyl chloride		
	VOC, unspecified		
	Zinc		

Cut-off at 1%; Different LCIA midpoint methods and inventories considered; EU-1 refers to inventory for Europe from Laurent et al. (2011); EU-2a refers to inventory for Europe from Sleeswijk et al. (2008); EU-2b refers to inventory for Europe from Sleeswijk et al. (2008), revised by the authors in 2010 (Sleeswijk AW (2010) Personal communication)

^a PAH polycyclic aromatic hydrocarbons (inventoried as benzo[a]pyrene or B[a]P)

In addition to its main purpose to facilitate the calculation of normalization references in other regions, the presented list could also help refining the USEtox™ characterization factors. All listed metals are currently flagged as interim in the model, and three organic substances, benzo(a)pyrene, dioxins (as 2,3,7,8-TCDD), and decabromophenyl ether, are also associated with at least one of their CFs flagged as interim. Table 5 thus suggests directing the focus towards getting reliable characterization factors for those substances.

Finally, it shall be noted that the provided list is not exhaustive. Although it relies on the most recent studies of the past decade, it still originates from inventories, whose chemical coverage is far from being complete. Several substances are thus expected to be missing either because of the incompleteness in inventories for Australia, Europe, and North America, or because other chemicals are simply not emitted any longer in these regions and are hence absent from the associated emission inventories. Additionally, the identified chemicals are also dependent on uncertainties related to the characterization factors, which might raise the significance of certain substances while decreasing others' (see Section 6.1).

6 Uncertainty analysis

6.1 USEtox™ model

Although uncertainty and variability have not yet been quantified explicitly for parameters in USEtox™, Rosenbaum et al. (2008) observed a variation of about three orders of magnitude among different models for human health impacts and two orders of magnitude for aquatic ecotoxicity impacts. These ranges essentially reflect variation between models as the substance parameters and most model parameters were identical among models. Furthermore, these observations have been made for chemicals that are characterized with recommended factors, i.e., substances for which USEtox™ is deemed well suited to model their impact potentials. Model uncertainties for interim characterization factors (e.g., for metals, dissociating, and amphiphilic organic and many inorganic chemicals) can be expected to be even larger since USEtox™ is best suited to model non-dissociating and non-amphiphilic organic substances. The speciation that occurs for metals and dissociating organic substances is thus not considered in the present version of the model. It should also be noted that uncertainties do not necessarily add up to an overall uncertainty. Even if parameter uncertainty can be

Table 4 Top contributing substances in the calculation of normalization references for North America and Australia

USEtox™ (NA) The present study	IMPACT2002+ (NA) Lautier et al. (2010)	TRACI (NA) Lautier et al. (2010)	EUSES (AU) Lundie et al. (2007)
Acrolein	Aluminum	Acrolein	Arsenic
Antimony	Arsenic	Arsenic	Atrazine
Arsenic	Atrazine	Azinphos-methyl	Cadmium
Atrazine	Benzene	Benzene	Cobalt
Barium	Benzo(a)pyrene	Benzo(a)pyrene	Copper
Cadmium	Chromium	Benzo(k)fluoranthene	Fluoride
Chlorothalonil	Copper	Cadmium	Formaldehyde
Chlorpyrifos	Decabromophenyl ether	Chlorpyrifos	Lead
Chromium	Dioxins	Copper	Manganese
Chromium VI	Lead	Cypermethrin	Mercury
Copper	Nickel	Deltamethrin	Metham sodium
Cyanazine	Zinc	Dioxins	Selenium
Cyfluthrin		Disulfoton	Simazine
Formaldehyde		Endosulfan	
λ-cyhalothrin		Glyphosate	
Lead		Hydrogen cyanide	
Mercury		Lead	
Nickel		Malathion	
Phosmet		Mercury	
Profenofos		Metolachlor	
Propanil		Parathion-ethyl	
Terbufos		Parathion-methyl	
Vanadium		Trichlorfon	
Zinc		Trifluralin	

Cut-off at 1% (except for Australia, for which distributions are not completely detailed); Different LCIA midpoint methods and inventories considered; NA refers to inventory for North America from Lautier et al. (2010); AU refers to inventory for Australia from Lundie et al. (2007).

high for a given substance and still comes on top of the aforementioned variability between models and thus contributes to overall uncertainty of the substance's USEtox™ characterization factors, it might not significantly increase overall uncertainty. Although the overall methodological uncertainty was not quantified here, it

may lead to changes in the contribution analyses of the calculated normalization references (Table 2). However, it is not expected to impact significantly on the high priority list established in Table 5, since USEtox™-based results only account for two out of the eight sets of the contribution analyses used as input data.

Table 5 High priority list of substances to inventory, based on previous studies

Organic substances	Occurrences	Metals	Occurrences
Atrazine	5	Copper	8
Chlorpyrifos	5	Lead	8
Benz(a)pyrene	4	Arsenic	7
Benzene	4	Cadmium	6
Cypermethrin	4	Mercury	6
Dioxins	4	Zinc	6
Formaldehyde	4	Chromium ^a	4
Acrolein	3	Nickel	4
Chlorothalonil	2	Antimony	2
Cyfluthrin	2	Barium	2
Decabromophenyl ether	2	Manganese	2
Parathion-methyl	2	Selenium	2
Terbufos	2	Vanadium	2

Substances with more than two occurrences in Tables 3 and 4; maximum number of occurrences is eight (i.e., substance appearing in all contribution analyses of Table 3)

^aChromium includes chromium (+VI)

6.2 Inventory analysis

The two inventories used in the present study are both associated with large errors due to insufficient coverage of substances as well as with uncertainties associated with extrapolations performed to build the emission inventories. These uncertainties will not be discussed here in details and the reader is thus referred to Laurent et al. (2011) and Lautier et al. (2010) for further information.

Focus was put on the influence of the inventory uncertainties on the calculated normalization references for Europe and North America. A sensitivity analysis was conducted, where inventory features that were identified as most influential in Laurent et al. (2011) and Lautier et al. (2010) were changed within their reasonable ranges, and variations on the normalization references were quantified. The resulting overall variability ranges for both sets of normalization references are shown in Table 6.

Because the influence of missing emission data could not be quantified, most scenarios of sensitivity analysis presented in Table 6 are limited to changes in existing inventory features, i.e. no emissions of substances initially not covered were added. However, even without trying to estimate the influence of the unknown omissions, the obtained variability ranges point to a trend of underestimation of the normalization references as ranges mainly stretch toward positive values. This finding suggests that

inventories are incompletely built even with regard to the substances they already encompass, i.e. the actual chemical emissions that are covered are only inventoried partially. Analyzing the variability ranges of Table 6, the inventory for North America seems to be more impacted than European inventory by this form of incompleteness.

High sensitivities in all normalization references are obtained when changes are assigned to metal emissions, with a variability ranging within one order of magnitude for Europe and scaling up to more than one order of magnitude for North America (freshwater ecotoxicity and non-carcinogenic human toxicity). These observations illustrate the strong dependence of the normalization references on metals, for which emission data are either poorly reported or insufficiently transparent (see Section 2). They also document the necessity to better inventory metal emissions along with the required refinement in the characterization model of metals expressed in Section 6.1. In both contexts, the focus should be directed towards the heavy metals identified in Table 5.

7 Conclusions

Normalization references for Europe and North America were calculated to be applied together with the USEtox™ characterization factors. In situations where the product life cycle stretches over other regions, the LCA practitioner is

Table 6 Sensitivity analysis on normalization references (inventory settings)

	Influence on normalization references			
	Freshwater ecotoxicity		Human toxicity (cancer)	Human toxicity (non-cancer)
	PestLCI	Pest-to-soil		
Inventory feature—Europe				
Heavy metals to soil based on Sleeswijk et al. (2008) ^{a,b}	+48%	+28%	+169%	+347%
Dioxins to water included ^b	+0%	+0%	+0%	+0%
Heavy metals to freshwater based on Sleeswijk et al. (2008) ^{a,b}	−67%	−39%	+31%	−18%
Variability ranges of normalization references—Europe	−67% < +48%	−39% < +28%	0% < +200%	−18% < +347%
Inventory feature—North America				
Addition of metals to agricultural soil in Canada based on extrapolations from Sleeswijk et al. (2008)	+47%	+16%	+52%	+648%
Inclusion of metal compounds in US emission inventory	+1484%	+516%	+335%	+1131%
Releases of certain metals to water based on Ecoinvent emission factors from energy production in US ^c	+34%	+10%	+9%	+71%
Variability ranges of normalization references—North America	0% < +1565%	0% < +542%	0% < +396%	0% < +1850%

^a Releases of heavy metals to freshwater and soil from Sleeswijk et al. rely on different data sources and inventory assumptions from the ones used in the present study; further information is available in Laurent et al. (2011) and Sleeswijk et al. (2008, 2010 Personal communication). Here, for the sensitivity analysis, emission data from the present study were substituted with the data from Sleeswijk AW (2010 Personal communication)

^b Sleeswijk AW (2010) Personal communication. The inventory from Sleeswijk et al. (2008) was revised by the authors in 2010

^c Specific metals include thallium, arsenic, barium, strontium, iron, nickel, antimony, and chromium (+VI)

advised to use as a default the European set of normalization references, which is deemed to be less uncertain than the one for North America. However, the LCA practitioner is also strongly advocated to consider the uncertainties that have been identified in the present study since they have the potential to influence the resulting normalized environmental profile. In that context, substantial uncertainties are demonstrated to stem from metals because of their overall dominance in all three USEtox™ impact categories and the interim status of their characterization factors as well as from the relatively poor emission data coverage for many organic substances that could contribute to the normalization references.

Guidance is provided to address future updates in the normalization references at both inventory and characterization levels. A list of priority substances was determined to highlight where to concentrate efforts when refining CFs or when collecting emission data. This list can thus not only serve as a framework for updating normalization references for Europe and North America but can also constitute a baseline for expanding normalization to other regions (i.e. continents) or to the world, which is deemed necessary due to the global scope addressed by the USEtox™ model and by any typical product LCA.

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